

LOWER CRITICAL FIELD MEASUREMENTS IN  
YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6+x</sub> SINGLE CRYSTALS

D.L. Kaiser, L.J. Swartzendruber, F.W. Gayle and L.H. Bennett  
Materials Science and Engineering Laboratory  
National Institute of Standards and Technology  
Gaithersburg, MD 20899

The temperature dependence of the lower critical field in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6+x</sub> single crystals has been determined by magnetization measurements with the applied field parallel and perpendicular to the c-axis. Results were compared with data from the literature and fitted to Ginzberg-Landau equations by assuming a linear dependence of the parameter  $\kappa$  on temperature. A value of  $7 \pm 2$  kOe was estimated for the thermodynamic critical field at  $T = 0$  by comparison of calculated  $H_{c2}$  values with experimental data from the literature.

## INTRODUCTION

The lower critical field ( $H_{c1}$ ) of the high temperature superconductor YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6+x</sub> (YBCO) is an intrinsic material property which depends upon temperature and crystallographic orientation. Accurate measurement of  $H_{c1}$  is complicated by flux pinning and edge effects (as illustrated in Fig. 1) and by uncertainty in the demagnetizing factor. Consequently, early reported  $H_{c1}$  values in single crystals<sup>1-5</sup> were up to an order of magnitude larger than later values.<sup>6-12</sup>

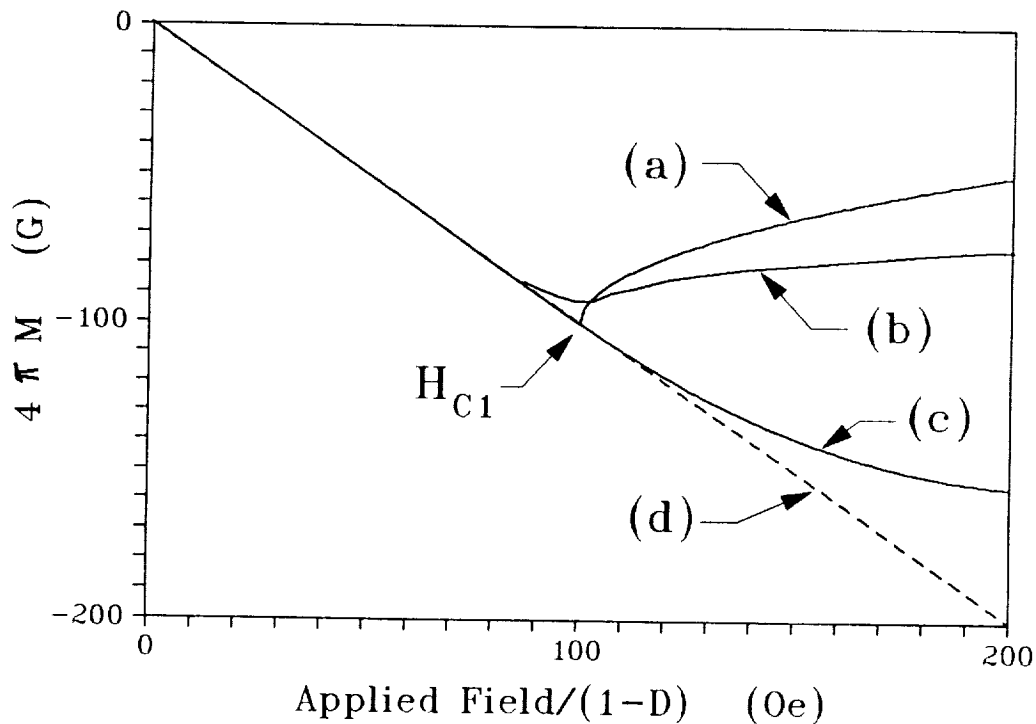
In this study,  $H_{c1}$  measurements were made on twinned and detwinned crystals and the data were compared with results of previous investigations for orientations with the applied field parallel and perpendicular to the c-axis of the crystal. The experimental data were fitted to Ginzberg-Landau equations for the dependence of  $H_{c1}$  on temperature and upper critical field ( $H_{c2}$ ) values were calculated for the two orientations of interest.

## EXPERIMENTAL PROCEDURE

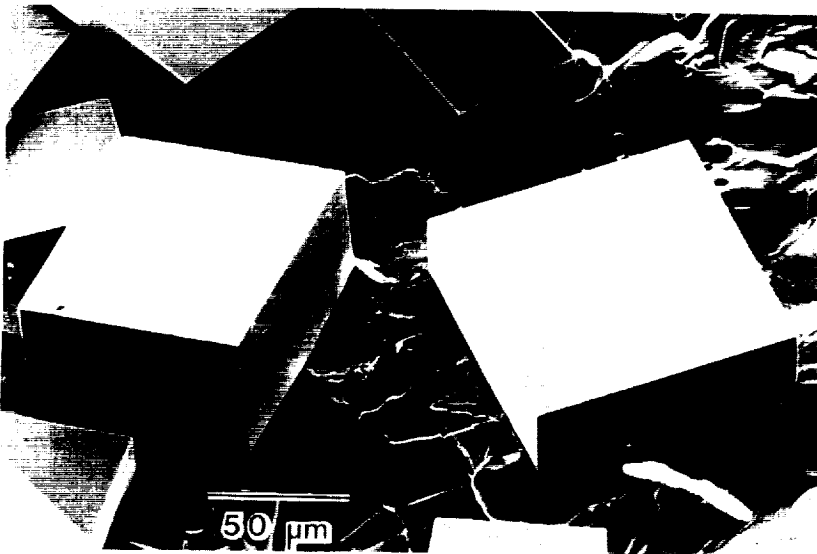
The YBCO crystals used in the present study (Fig. 2) were grown from Y-Ba-Cu-O

melts<sup>13</sup> and subsequently annealed in oxygen gas at 420°C for 80 h to obtain superconducting transition temperatures  $T_c > 90$  K. (Thus, the oxygen content  $6+x > 6.85$ <sup>14</sup>). Two crystals were selected for measurement. The first crystal (AN3-5) exhibited characteristic (110) twin planes and was nearly cubic with dimensions  $120 \times 135 \times 120 \mu\text{m}^3$  (c-dimension =  $120 \mu\text{m}$ ). Due to the cubic morphology, the demagnetizing factors were nearly identical in all three dimensions. The second crystal (AN9-5) was fully detwinned via a thermo-mechanical process developed in our laboratory<sup>15</sup> and had dimensions  $a \times b \times c = 200 \times 250 \times 100 \mu\text{m}^3$ .

Magnetic measurements were made using a superconducting quantum interference device (SQUID) magnetometer, with the c-axis of the crystal aligned either perpendicular or parallel to the applied field  $H$ . The crystal was first cooled in zero field to a predetermined temperature and the magnetization was then measured as the applied field was increased to a value in excess of  $H_{c1}$ . For temperatures greater than about 60 K, sharp breaks from linearity in the  $M$  vs.  $H$  curves were observed for both crystals, making the estimation of  $H_{c1}$  relatively precise. Below 60 K, the



**Fig. 1.** Hypothetical magnetization curves after cooling in zero field for a sample: (a) at equilibrium (i.e., an ellipsoid with no flux pinning); (b) with some pinning and edge effects; (c) with strong pinning; and (d) with perfect diamagnetic character (i.e., magnetization is proportional to the applied field after correction for the demagnetizing factor,  $D$ ). For the equilibrium case (a), there is a well-defined, sharp break at  $H_{c1}$ , which allows for an accurate determination of  $H_{c1}$ . Pinning and edge effects (b and c) make it difficult to estimate the true  $H_{c1}$ . In case (b), the observed onset occurs at applied fields below  $H_{c1}$  due to flux penetration at the sharp edges and corners of the crystal. In case (c), pinning causes a gradual departure from linearity, making the estimate of  $H_{c1}$  less accurate.



**Fig. 2.** Scanning electron micrograph of as-grown YBCO single crystals. The smallest dimension of a crystal generally lies along the  $c$ -axis of the unit cell.

departure from linearity was more gradual and  $H_{c1}$  was estimated from the initial point of departure from linearity.

## RESULTS AND DISCUSSION

Temperature-dependent  $H_{c1}$  data from the present study and previous investigations<sup>4,6-8,10-12</sup> for  $H \parallel c$  and  $H \perp c$  are presented in Fig. 3. The earliest reported  $H_{c1}$  values<sup>1-3,5</sup> were erroneously high due to difficulties in defining  $H_{c1}$  and are not included in the two plots. The curves shown in each plot were obtained by fitting all displayed data points using the Ginzberg-Landau equation<sup>16</sup>

$$H_{c1} = H_c (\ln \kappa + 0.08) / \sqrt{2} \kappa, \quad (1)$$

where  $H_c$  is the thermodynamic critical field at temperature  $T$  as given by

$$H_c = H_{c0} (1 - t^2). \quad (2)$$

Here  $H_{c0}$  is  $H_c$  at  $T = 0$ ,  $t$  is the reduced temperature  $T/T_c$  and  $\kappa$  is the Ginzberg-Landau parameter (the ratio of the penetration depth  $\lambda$  to the coherence length  $\xi$ ). Data for both  $H \parallel c$  and  $H \perp c$  can be well-fitted by assuming that  $\kappa$  varies linearly with temperature:

$$\kappa = a + bt. \quad (3)$$

Good fits of Eq. (1) may be obtained for a wide range of  $H_{c0}$  values, leading to a wide range of values for the parameters  $a$  and  $b$  in Eq. (3). The range of permissible  $H_{c0}$  values is limited considerably by requiring that  $H_{c2}$  values calculated from a second Ginzberg-Landau equation

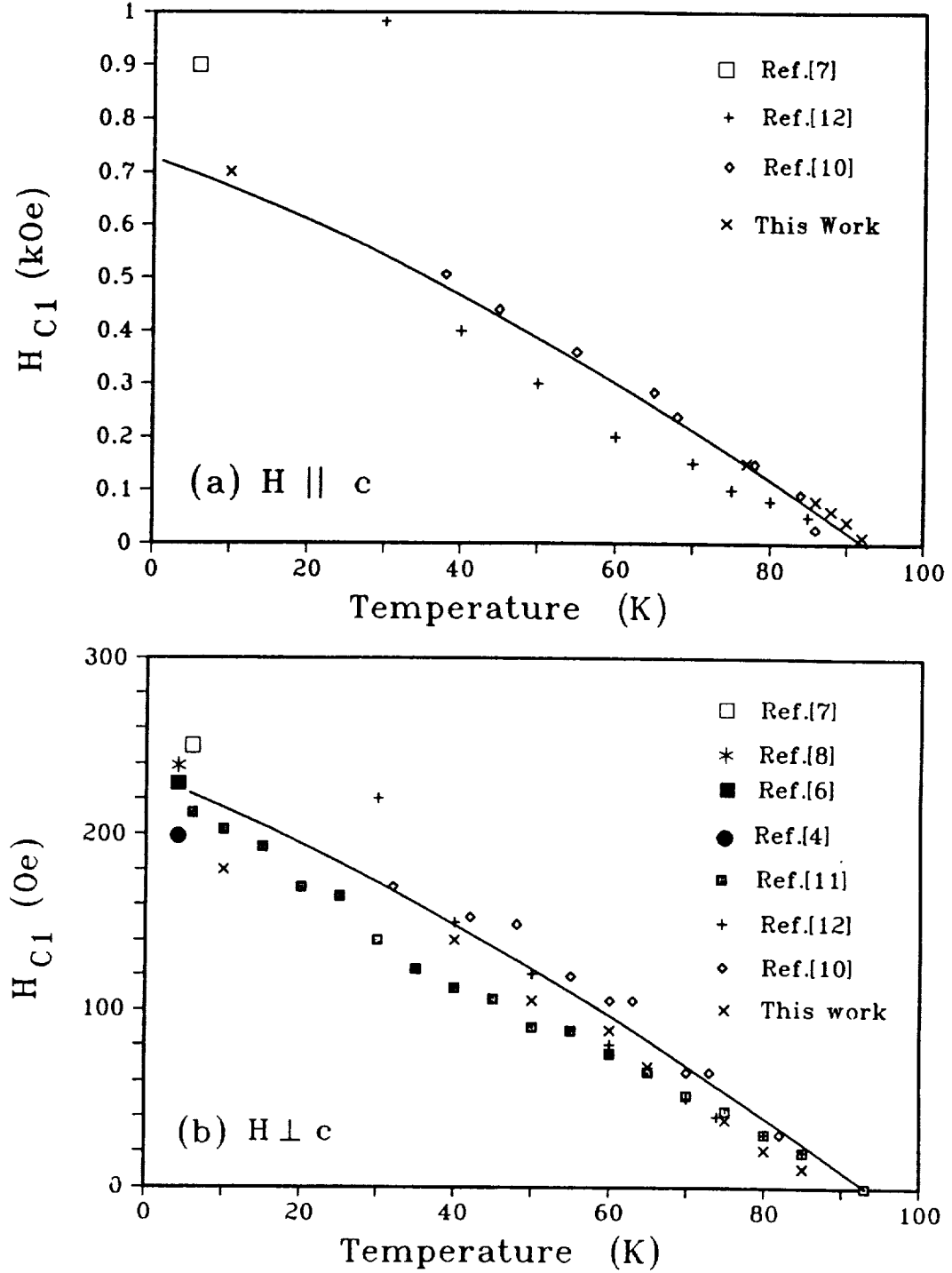
$$H_{c2} = \sqrt{2} \kappa H_c \quad (4)$$

be in reasonable agreement with experimental  $H_{c2}$  data from the literature. This comparative analysis yields a value of 7 kOe for  $H_{c0}$ ,  $\kappa = 100 + 85t$  for  $H \perp c$  and  $\kappa = 22 + 22t$  for  $H \parallel c$ . The

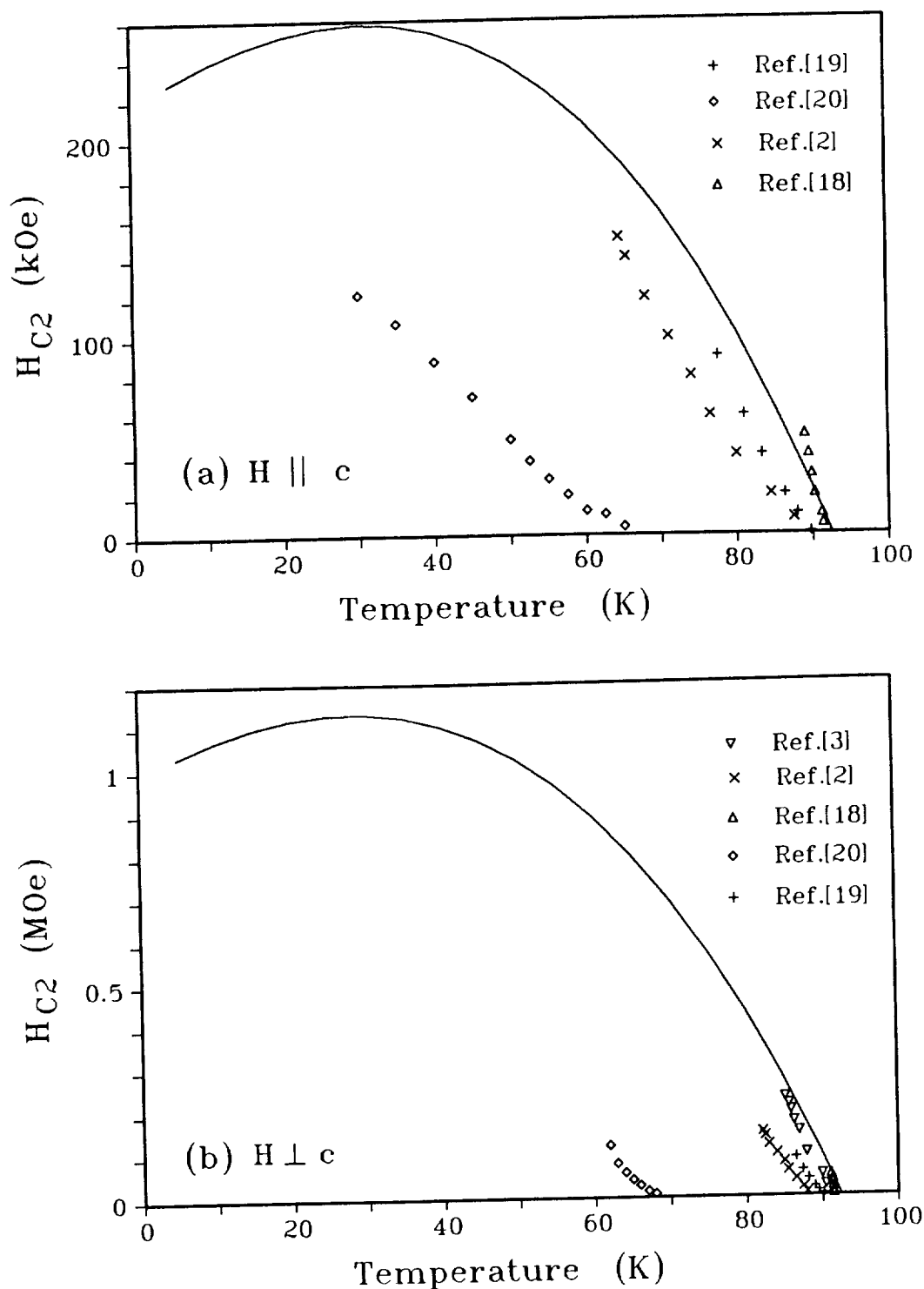
calculated curves and data for  $H_{c2}$  are compared in Fig. 4. Considering the large uncertainties in the experimental  $H_{c2}$  data and the obvious differences in  $T_c$  for samples from different studies, agreement of the calculated curves with the data is a matter of judgement. The maxima seen in the calculated  $H_{c2}$  curves are unphysical, indicating that the linear form used for  $\kappa$  should be modified, e.g., by the addition of a quadratic term in  $t$ . However, the large uncertainty in the  $H_{c0}$  value used here ( $7 \pm 2$  kOe) does not justify such an additional term. Considering the large variations in the experimental  $H_{c2}$  data, our  $H_{c0}$  value is in reasonable agreement with the value of 10 kOe estimated by Worthington et al.<sup>6</sup> The resulting uncertainties in  $\kappa$  values calculated from our equations are also of order  $\pm 30\%$ .

Our  $H_{c1}$  data for the detwinned crystal AN9-5 shown in Fig. 3a ( $H \parallel c$ ) are in good agreement with the data of Krusin-Elbaum et al.<sup>10</sup> for twinned crystals. This result indicates that twin boundaries have only a small effect on  $H_{c1}$ , as noted in our earlier investigation.<sup>17</sup> Our  $H_{c1}$  data for the twinned crystal AN3-5 shown in Fig. 3b ( $H \perp c$ ) are in reasonable agreement with the data from previous investigations.<sup>4,6-8,10-12</sup> Anisotropy in  $H_{c1}$  ( $H_{c1} \parallel c/H_{c1} \perp c$ ) as calculated from the two curves in Fig. 3 was  $3.1 \pm 0.1$  for  $10 \text{ K} < T < 80 \text{ K}$ .

The calculated  $H_{c2}$  values for  $H \parallel c$  (Fig. 4a) are in reasonable agreement with the data of Welp et al.<sup>18</sup> near the superconducting transition and follow the general trend of the data of Iye et al.<sup>19</sup> and Worthington et al.<sup>2</sup>. For  $H \perp c$  (Fig. 4b) the calculated values at high temperature show good agreement with the experimental data of Gallagher et al.<sup>3</sup> and Welp et al.<sup>18</sup> Anisotropy in  $H_{c2}$  ( $H_{c2} \perp c/H_{c2} \parallel c$ ) as calculated from the two curves in Fig. 4 was  $4.3 \pm 0.2$  for  $10 \text{ K} < T < 80 \text{ K}$ .



**Fig. 3.** Temperature-dependent  $H_{c1}$  data for (a)  $H \parallel c$  (strong pinning) and (b)  $H \perp c$  (weak pinning). Our data are for (a) the detwinned crystal AN9-5 and (b) the twinned crystal AN3-5. The curves were generated by fitting the data to the Ginzberg-Landau equation for  $H_{c1}$  and temperature-dependent  $\kappa$  equations given by (a)  $\kappa = 22 + 22t$ , and (b)  $\kappa = 100 + 85t$  ( $t = T/T_c$ ).



**Fig. 4.** Temperature-dependent  $H_{c2}$  curve (solid line) calculated from the Ginzberg-Landau equation for  $H_{c2}$  and the temperature-dependent  $\kappa$  equation for (a)  $H \parallel c$  and (b)  $H \perp c$ . Experimental  $H_{c2}$  data from the literature are shown for comparison. The extrapolations to lower temperatures ( $T < 60$  K) are unreliable and the maxima in the curves probably do not exist.

## CONCLUSIONS

Temperature-dependent  $H_{c1}$  results were determined from magnetization measurements on detwinned and twinned single crystals of YBCO for  $H \parallel c$  and  $H \perp c$ . The results from the present study and previous investigations for each orientation were fitted to Ginzberg-Landau equations assuming a linear temperature dependence for the parameter  $\kappa$ .  $H_{c2}$  values calculated from the Ginzberg-Landau equation and the temperature-dependent  $\kappa$  relations were in reasonable agreement with experimental  $H_{c2}$  data from the literature near the superconducting transition temperature. Values of  $H_{c0} = 7 \pm 2$  kOe,  $\kappa = 100 + 85t$  for  $H \perp c$ , and  $\kappa = 22 + 22t$  for  $H \parallel c$  were estimated from the analysis.

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